Tuning of Hidden Order and Superconductivity in URu_2Si_2 by Applied Pressure and Re Substitution

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ABSTRACT

Single crystals of $URu_{2-x}Re_xSi_2$ have been grown via the Czochralski technique. Detailed electrical transport studies under pressure on single crystals of URu_2Si_2 confirm that the zero-temperature critical field is suppressed smoothly towards an extrapolated critical pressure of 15 kbar, which also corresponds to the accepted critical pressure of the hidden order phase. Improving on previous work on polycrystalline samples, studies of single crystals of $URu_{2-x}Re_xSi_2$ have provided more precise tracking of the suppression of both the hidden order phase at low doping and the ferromagnetic phase at intermediate Re concentrations.

INTRODUCTION

Since the discovery that below about 17 K the heavy-fermion superconductor ($T_c \approx$ 1.5 K) URu_2Si_2 exhibits an ordered phase with an uncertain order parameter [1-3], this hidden order (HO) phase has been the subject of much experimental and theoretical research. At the core of the mystery is a discrepancy between the large size of a jump in the specific heat at the ordering temperature, and the small size of the staggered antiferromagnetic (AFM) moment observed by neutron scattering in the ordered phase [4]. The specific heat anomaly is consistent with the formation of a partial gap over the Fermi surface [2], although many other scenarios have been proposed [5]. The disagreement in magnitude between specific heat and staggered moment has led some researchers to believe that the small observed moment is either coupled to a different primary order parameter, or arises from a competing magnetic phase.

An often-useful experimental approach to the identification of ordered phases is the modification of a material via the application of external magnetic field, external pressure, or chemical substitution. The HO phase of URu_2Si_2 persists to remarkably high magnetic fields of about 37 T [6], but it is much more sensitive to the application of relatively low pressures and chemical substitution onto the Ru site. Early studies of transition metal substitution for Ru in polycrystalline samples found that both the HO/AFM and superconducting phases are suppressed at substituent levels smaller than 10% [7-9]. More recently, it was found that non-Fermi liquid behavior, often associated with a quantum critical point, at which an ordering temperature is driven to 0 K, persists well into the ferromagnetic ordered phase that exists at moderate Re concentration in $URu_{2-x}Re_xSi_2$, a very unusual occurrence [10]. Early pressure studies on URu_2Si_2 showed that applied pressure suppresses superconductivity, while the HO/AFM transition temperature is increased [11] and the measured moment grows substantially [12]. However, there still exist disagreements about the relationship between HO, AFM, and superconductivity as well as the general temperature-pressure phase diagram [12-16]. To more precisely map the pressure dependence of the hidden order and superconducting phases, electrical resistivity measurements on single crystals of URu_2Si_2 are being performed as a

function of temperature, magnetic field, and pressure. To study the interaction between the various phases and search for effects of quantum criticality, the physical properties of single crystals of $URu_{2-x}Re_xSi_2$ are being measured as a function of temperature, magnetic field, and pressure. Preliminary results of these measurements are presented herein.

EXPERIMENTAL DETAILS

Single crystals of $URu_{2-x}Re_xSi_2$ were pulled via the Czochralski technique in a tri-arc furnace and annealed in an Ar atmosphere at 900 ^{o}C for 7 days. Samples were oriented by the Laue method and prepared for measurement by cutting with a diamond wheel saw and spark erosion. Powder x-ray diffraction indicated single-phase material, while clean Laue patterns confirmed well-oriented crystals. Electrical resistivity ρ measurements as a function of temperature T under applied pressure P were performed in a hydrostatic Be-Cu piston-cylinder clamp using a 1:1 solution of isoamyl alcohol and n-pentane as the pressure-transmitting medium. The cells were measured in an Oxford Kelvinox MX100 dilution refrigerator equipped with a magnet capable of fields H up to 9 T. Pressure was inferred from the superconducting transition of Pb. Measurements of magnetization M were made using a Quantum Design Magnetic Properties Measurement System (MPMS). Measurements of $M(T,H)$ and $\rho(T,H)$ were done with H parallel to the c-axis, and current was parallel to the a-axis for all ρ measurements.

DISCUSSION

The electrical resistivity of URu_2Si_2 in the vicinity of the HO/AFM transition at different applied pressures is shown in Figure 1a. The HO/AFM transition temperature T_o, defined as the inflection point in $\rho(T)$, clearly increases with P. It is also evident that the height of the peak below the transition is suppressed as P increases, while the peak width increases. This may be due to the development of pressure inhomogeneity at higher pressures, to which URu_2Si_2 is sensitive, or may reflect a change in scattering as the ordered phase evolves from HO to AFM.

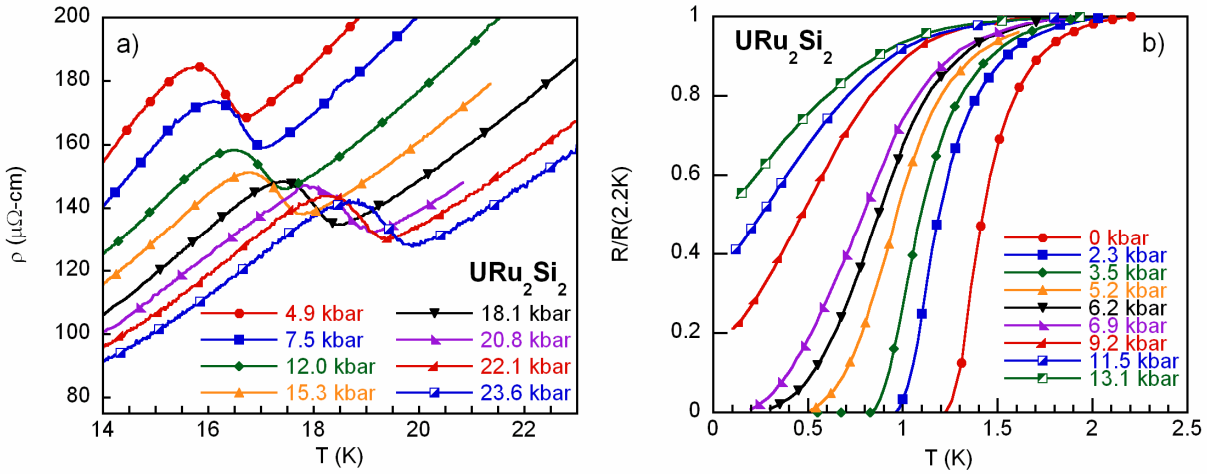

Figure 1. a) Electrical resistivity $\rho(T)$ of URu_2Si_2 at various pressures. The HO/AFM transition temperature increases with applied pressure. b) Low temperature $\rho(T)$ at various pressures, demonstrating the suppression of the superconducting transition by applied pressure.

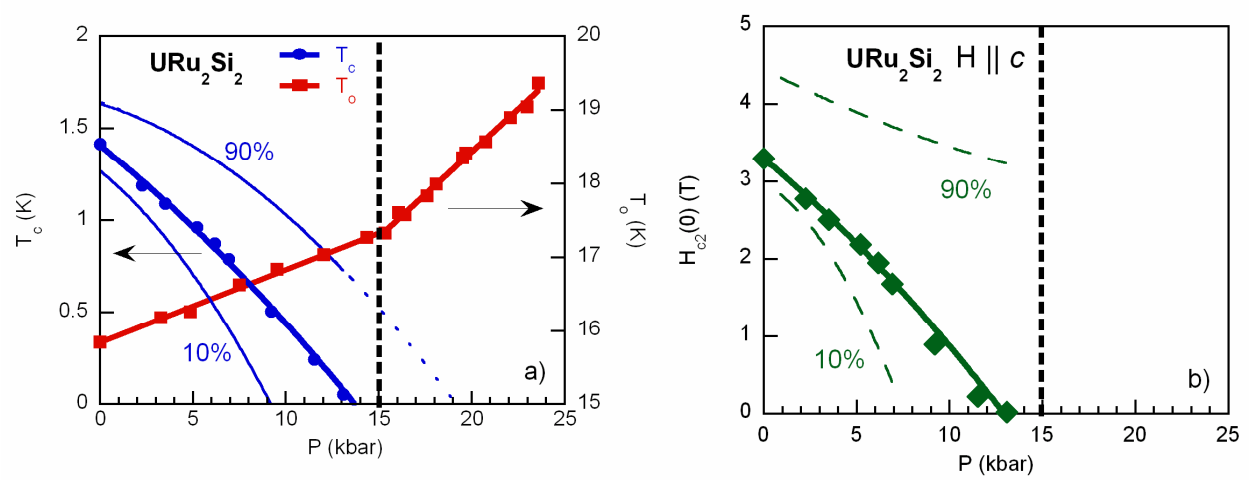

Figure 2. a) Pressure dependence of T_c (left axis), HO/AFM transition temperature T_o (right axis), and b) zero-field upper critical field $H_{c2}(0)$. $\partial T_o/\partial P$ roughly doubles at 15 kbar (dashed vertical line), where T_c and $H_{c2}(0)$ extrapolate to 0 K. Envelopes correspond to 10% and 90% of the $\rho(T,H)$ superconducting transition.

Figure 1b illustrates the suppression of the superconducting transition temperature T_c as P increases, with $H = 0$. The somewhat wide superconducting transition in $\rho(T)$, typical of this compound, widens with pressure, complicating the identification of T_c. Here T_c is defined as the temperature at which the value of ρ is 50% of its normal state low-T value. The values of the upper critical field $H_{c2}(0)$ are determined directly by plotting these $\rho(T)$ transitions as a function of H. Figure 2 summarizes the P dependence of T_o, T_c, and $H_{c2}(0)$, along with envelopes that delineate the temperatures at which $\rho(T)$ equals 10% and 90% of the normal state value. Both T_c and $H_{c2}(0)$ extrapolate to 0 K at a critical pressure P_c of about 15 kbar. Although the value of T_o steadily increases with P, $\partial T_o/\partial P$ changes from 0.1 K/bar to 0.23 K/kbar at P_c, suggesting a relationship between the disappearance of superconductivity and a change in the HO/AFM phase. In a scenario where HO and AFM phases both compete for sample volume, it is plausible that the superconductivity is the ground state of the HO phase and disappears when HO is no longer present. The difference between this estimate of P_c and those of recent reports [14,15] may be due to sample dependence, measurement technique, or differing definitions of transition temperatures. Regardless, the data from this work are difficult to reconcile with the existence of a first-order phase boundary, demarcating small-moment and large-moment phases, that terminates at 5 kbar at 0 K [14,15], as no qualitative change is observed in the superconductivity around that pressure, even when transition width is accounted for.

In contrast to applied pressure, substituting Re for Ru in $URu_{2-x}Re_xSi_2$ suppresses both superconductivity and the HO/AFM phase. Whether HO converts to AFM with chemical substitution has not been assessed. From ρ(T) data of the single crystals, it is possible to track the value of T_o up to $x = 0.10$ (Figure 3a), while for polycrystals this was only possible using specific heat data. Substitution of Re appears to broaden the peak below T_o and above $x = 0.10$, T_o is no longer distinguishable. Compared to polycrystals, T_o in single crystals is lower, perhaps due to a more homogeneous Re distribution in the single crystals. Two possible explanations for the suppression of T_o with increased Re concentration are that substituting Re for Ru induces a lattice expansion, and that Re substitution removes electrons from the conduction band at the

Fermi energy. It is not clear which mechanism should dominate, but it is noteworthy that Re doping barely changes the URu_2Si_2 lattice at the low concentrations in question [7].

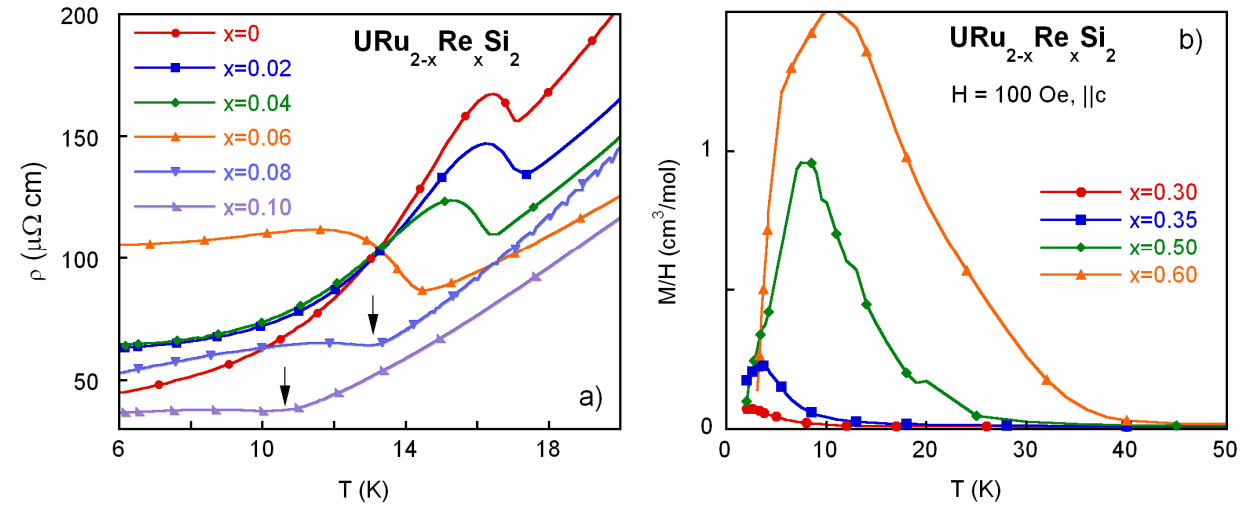

Figure 3. a) Electrical resistivity $\rho(T)$ of $URu_{2-x}Re_xSi_2$ in the vicinity of the HO/AFM transition T_o. The data have been vertically offset for clarity. b) Magnetic susceptibility $\chi(T)$ in the ferromagnetic region showing the increase in the ferromagnetic transition temperature.

To more accurately identify the Re concentration range at which long range ferromagnetism is suppressed, $M(T,H)$ measurements on samples at intermediate Re concentration were performed (Figure 3b). In these examples, a clear maximum in the dc magnetic susceptibility $\chi \approx M/H$, measured in a field of 100 Oe, correlates with the onset of ferromagnetic order. Although this feature is unusual for a ferromagnet, mean-field Arrott analysis supports the correspondence between the maximum in $\chi(T)$ and a ferromagnetic transition at $x = 0.5$. Furthermore, long-range ferromagnetism was confirmed by neutron scattering for $x = 0.8$ [17].

Despite the changes in ground state induced by Re substitution, the high-T ($T > 120$ K) paramagnetic moment, as determined from Curie-Weiss law fits, does not change with Re doping. Similarly, the $\rho(T)$ data are also independent of x for $T > 80$ K, indicating that Re doping only changes the properties of URu_2Si_2 below its coherence temperature. The Curie-Weiss temperatures determined from the $\chi(T)$ fits remain constant with values of about –120 K for $x < 0.35$, suggesting that antiferromagnetic correlations persist at high T, independent of Re substitution. However, for $x > 0.35$, the Curie-Weiss temperature increases to a value of –75 K by $x = 0.6$, tracking the evolution of the low-T ferromagnetic phase.

Plotting the suppression of both the HO/AFM and ferromagnetic phases yields the phase diagram shown in Figure 4. The phase boundary of the HO/AFM transition extrapolates to $x \approx 0.12$, although it is unclear whether the phase boundary extends to zero temperature or simply terminates because the last finite-temperature transition is seen at the relatively high temperature of 10 K in $x = 0.1$. Extrapolating the ferromagnetic phase boundary to 0 K yields a critical concentration x_c of approximately $x = 0.2$, in rough agreement with the value $x_c = 0.3$ published earlier for polycrystalline samples. One motivation for the identification of $x = 0.3$ with criticality in the polycrystalline samples was that the magnitude of the non-Fermi liquid like low-T logarithmic divergence in the specific heat divided by temperature C/T was greatest at that

concentration. To confirm this behavior in single crystals, specific heat measurements are currently being performed, along with transport and inelastic neutron scattering measurements, to test for the existence of non-Fermi liquid behavior at these concentrations. Single crystals are also being measured to study the anisotropy of $URu_{2-x}Re_xSi_2$ using transport and magnetization probes.

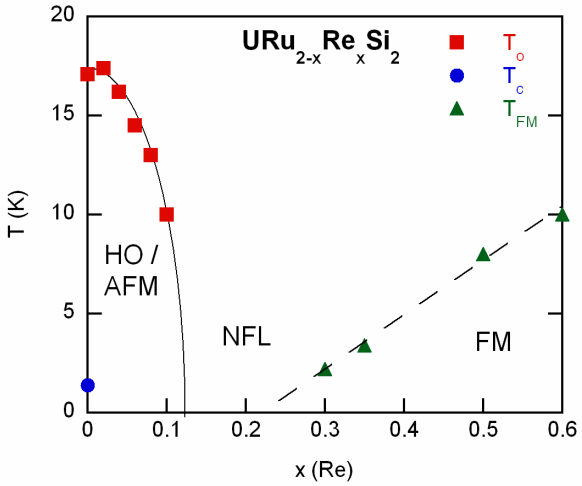

Figure 4. Phase diagram for $URu_{2-x}Re_xSi_2$. The HO/AFM and ferromagnetic (FM) phases are suppressed towards concentrations in the vicinity of $x = 0.2$. Non-Fermi liquid behavior has been observed at these and higher concentrations in polycrystalline samples, and is being investigated in single crystal samples.

CONCLUSIONS

The pressure and Re concentration dependence of the HO/AFM and superconducting phases of URu_2Si_2 are being studied. Under pressure, T_c and H_{c2} are suppressed smoothly towards a critical pressure of 15 kbar, the pressure at which the pressure dependence of the HO/AFM transition $\partial T_o/\partial P$ doubles, suggesting a correspondence between the two phenomena. Substitution of Re for Ru suppresses both the HO/AFM and superconducting phases, and leads to the emergence of ferromagnetic order at higher concentrations. The effects of both these tuning parameters continue to be investigated.

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